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DOCKET NO.: 240316US0XCONT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

Michele BORTOLOTTI et al.

: GROUP ART UNIT: 1714

SERIAL NO: 10/627,764

FILED: July 28, 2003

FOR: ELASTOMERIC COMPOSITION USEFUL AS TIRE TREADS

DECLARATION UNDER 37 C.F.R. 51.132COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

Now comes GIAN TOMMASO VIOLA who depose and state that.1. I am a graduate of CHEMISTRY and received myGRADUATE degree in the year 1974.2. I have been employed by POLITER (EUROPA) for 18 yearsas a RESEARCHER in the field of POLYMERS.

3. I am a graduate of _____ and received my

_____ degree in the year _____.

4. I have been employed by _____ for _____ years

as a _____ in the field of _____.

5. The following experiments were carried out and the resulting data are as follows:

I. EXAMPLE I

Styrene-butadiene copolymers A1, A2, A3 and A4 were prepared as follows:

8,000 g of an anhydrous cyclohexane-n-hexane mixture in a ratio of 9:1 by weight, 64 g of THF and 250 g of styrene and 750 g of butadiene were introduced into a 20 liter reactor equipped with a stirrer. The temperature of the mixture was brought to 40°C and 0.64 g of n-butyl lithium in cyclohexane was fed into the mixture. The beginning of polymerization was indicated by an increase in temperature. When a maximum temperature of about 80°C was reached, the solution was left under stirring for about 5 minutes. Then 0.6 g of diphenylcarbonate in hexane solution was added and the mixture was left to react under stirring for another 10 minutes, until the polymerization reaction was complete and unepoxidated reference copolymer A1 was produced.

A 2000 g portion of the polymeric solution A1 was transferred to a second reactor, where it was subjected to an epoxidation reaction by the addition of formic acid and hydrogen peroxide with a molar ratio with respect to the double bond equal to 15/15/100. The temperature of the polymeric solution, to which 21 g of formic acid was added, was raised to 70°C and 58.6 g of hydrogen peroxide (30% w/w) was added dropwise over a period of from 5 to 30 minutes. At the end of the addition the polymeric solution was maintained at about 70°C for a period of 1 to 5 hours. The epoxidation reaction was terminated by eliminating both the water and formic acid. The pH of the solution was adjusted to a value of about 7 and copolymer A2 was produced.

A 2,000 g portion of the polymeric solution A1 was mixed with 2.9 g of formic acid and the temperature was raised to about 70°C. 8.0 g of hydrogen peroxide (30% w/w) was added and the same procedure was carried out as disclosed above in the production of copolymer A2 to produce copolymer A3.

A 2,000 g portion of the polymeric solution A1 was mixed with 47 g of formic acid and the temperature was raised to about 70°C. 133.0 g of hydrogen peroxide (30% w/w) was

added and the same procedure was carried out as disclosed above in the production of copolymer A2 to produce copolymer A4.

0.3 phr of 2,6-di-*t*-butyl phenol was added to the polymeric solutions A1, A2, A3 and A4. The mixtures were coagulated with isopropyl alcohol and the coagulated products were dried in an oven at 60°C for 4 hours to produce dry copolymers A1, A2, A3, and A4.

II. EXAMPLE II

Using the procedures analogous to that set forth above in Example 1, three styrene-butadiene copolymers were prepared, the first being an unepoxidated reference copolymer A5.

A 2,000 g portion of the polymeric solution A5 was mixed with 10.0 g of formic acid and the temperature was raised to about 70°C. 26.5 g of hydrogen peroxide (30% w/w) was added and the same procedure was carried out, as discussed above in Example 1 to produce copolymer A6.

A 2,000 g portion of the polymeric solution A5 was mixed with 60.5 g of formic acid and the temperature was raised to about 70°C. 165.0 g of hydrogen peroxide (30% w/w) was added and the same procedure was carried out, as discussed above in Example 1 to produce copolymer A7.

The properties of copolymers A1-A7 are shown in Table 1, wherein % epoxidation refers to the molar percentage of epoxidated double bonds with respect to the mols of the initial diene double bonds.

TABLE 1

Copolymer	A1	A2	A3	A4	A5	A6	A7
styrene	25.1	25.0	25.0	24.8	25.1	24.9	25.0
Vinyl %	47.2	50.2	50.3	50.3	63.5	64.9	65.0
<Mw>	259300	254300	n.d.	238100	246800	239400	225000
<Mn>	209000	211000	n.d.	199000	191000	180000	171000
T _g	-35°C	-29°C	-35°C	-23°C	-21°C	-20°C	-18°C
Epoxidation %	0	5	0.68	11	0	2.27	14
ML _{1+100°C}	58	67	54	69	53	53	56

III. PREPARATION OF VULCANIZED PRODUCTS

100 parts styrene-butadiene copolymer A1, Cumaronic resin 2 phr, Silica BN3 53 phr, Carbon black N330 4.25 phr, bis[3-triethoxysilylpropyl] tetrasulfide (Si 69) 4.25 phr, ZnO 2.5 phr, Stearic acid 1.0 phr, Antioxidant 1.0 phr, Microcrystalline wax 1.0 phr, Aromatic oil 6.0 phr, N-cyclohexyl benzothiazolesulfenamide 1 phr, diphenylguanidine 1.5 phr and Sulfur 1.8 phr were mixed using an internal Banbury type laboratory mixer in 2-step mixing cycles: the first, for incorporating the charges and (Si 69), was carried out in a Banbury mixer operating to obtain discharge temperatures of between 140 and 160°C; the second, for the addition of the vulcanizing system, was carried out in an open mixer; the total mixing time being 9 minutes. Vulcanization was carried out in a press at 151°C for 60 minutes.

Copolymers A2-A7 were vulcanized in the same manner as copolymer A1 to produce vulcanized products M1-A1, M1-A2, M1-A3, M1-A4, M1-A5, M1-A6 and M1-A7.

The properties of the vulcanized products are shown in Table 2.

TABLE 2

Compound	M1-A1	M1-A2	M1-A3	M1-A4	M1-A5	M1-A6	M1-A7
100% Modulus (MPa)	4.5	5.3	4.4	6.5	4.2	4.4	5.8
200% Modulus (MPa)	8.9	11.3	9.3	13.7	10.2	11.2	13.1
Tensile strength (MPa)	16.3	17.5	18.3	17.1	17.0	17.5	18.2
Elongation at break (%)	332	282	349	163	294	282	143
Hardness (Shore A)	78	75	77	82	73	72	79
Abrasion loss (mm ³)	136	111	125	157	153	146	210
Tan δ 1Hz, 0.1% strain, 0°C	0.127	0.247	0.126	0.276	0.432	0.648	0.704
Tan δ 1Hz, 5% strain, 60°C	0.138	0.097	0.142	0.121	0.079	0.077	0.081
Tan δ 1Hz, 10% strain, 60°C	0.155	0.102	0.153	0.150	0.132	0.125	0.143
Tan δ 1Hz, 0.1% strain, 0°C/Tan δ 1Hz 5% strain, 60°C	0.92	2.55	0.89	2.28	5.47	8.42	8.69

IV. THE RESULTS OF THE EXPERIMENTS ABOVE ARE INTERPRETED AS FOLLOWS:

The data of Tables 1 and 2 should be divided and interpreted as two separate groups, the first group being copolymers A1-A4 and the second group being copolymers A5-A7. This is because the copolymers of the two groups differ in vinyl content, which is 47.2-50.5% for copolymers A1-A4 and 63.5-65.0% for copolymers A5-A7. The vinyl content of the copolymers influences the properties of the copolymers, in particular T_g (glass transition temperature) and, consequently, dynamic behavior and resistance to abrasion.

V. DISCUSSION OF THE RESULTS OF TESTS ON M1-A1 TO M1-A4

Tan δ 1 Hz, 0.1% strain, 0°C is considered a good measurement of the "wet grip" of a tire, while Tan δ 1 Hz, 5% strain, 60°C is a good measurement of the "rolling resistance" of a tire.

The ratio Tan δ 1 Hz, 0.1% strain, 0°C/Tan δ 1 Hz, 5% strain, 60°C is a significant ratio, because it represents a compromise between "wet grip" and "rolling resistance" and high values show an optimization of a dynamic performance of the composition making up the tire, assuming the mechanical properties of the tire are the same. As can be seen from Table 2 the ratio is highest for M1-A2 at 2.55, which represents copolymer A2 within the epoxidation % of the present claims at 5% epoxidation, as compared to M1-A1 at 0.92, which is the unepoxidated reference copolymer and M1-A3 and M1-A4 at 0.84 and 2.28, respectively, both of which are outside the epoxidation % of the present claims of 2.27-5%, copolymer A3 being 0.68% epoxidation and copolymer A4 being 11% epoxidation. Further, M1-A2 is superior in minimizing abrasion loss against M1-A1, M1-A3 and M1-A4 and M1-A2 is also superior in not demonstrating too high of a hardness against M1-A1, M1-A3 and M1-A4. The above results are especially significant between M1-A2, having a 5% epoxidation and within the range of the present claims with an abrasion loss of 111 and a Shore hardness of 75 as compared to M1-A4, having an 11% epoxidation, outside the range of the present claims with an abrasion loss of 157 and a Shore hardness of 82.

Comparing M1-A5, M1-A6 and M1-A7, it can be seen that M1-A6, which has an epoxidation % of 2.27, within the range of the present claims, is much better in the above-discussed ratio than M1-A5, which uses a copolymer with an epoxidation % of 0 and very close to but slightly less than M1-A7, which uses a copolymer with an epoxidation degree of 14%, outside the range of the present claims. However, M1-A6 is much superior in

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minimization of abrasion loss, as compared to M1-A5 and M1-A7 and is also superior in not demonstrating too high a hardness, as compared to M1-A5 and M1-A7. The results are especially significant when the abrasion loss of M1-A6 of 146 is compared to the abrasion loss of 210 for M1-A7 and the Shore hardness of M1-A6 of 72 is compared to that of M1-A7 of 79.

Therefore, it can be seen that the comparative data in Table 2 demonstrates superior results for compositions within the epoxidation percent range of the present claims, as compared to compositions of the prior art, which are outside the epoxidation percent range of the present claims.

6. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

7. Further deponents state not

John Thomas DeLoe
Signature

9/FEB/2004
Date

Signature

Date